Intrinsic catalytic activity of carbon nanotubes for electrocatalytic nitrate reduction to ammonia

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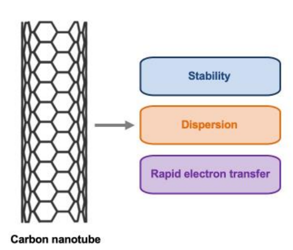
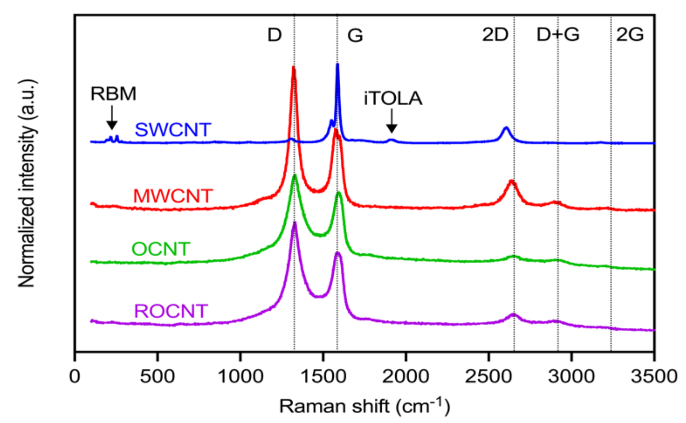
Introduction

Nitrate (NO_3^-) ions are among the most widespread water pollutants impacting surface water and groundwater on a global scale. Electrochemical NO_3^- reduction is a promising approach to remove nitrate contaminants by converting them to value-added fuels and chemical products, such as ammonia (NH_3). There is growing interest in developing active and selective electrocatalysts for converting NO_3^- to NH_3. Carbon nanomaterials, including carbon nanotubes (CNTs), have been demonstrated to have a wide application in catalysis due to their desirable properties. **This work investigates different types of CNTs as catalysts for the electroreduction of NO_3^- to NH_3.**

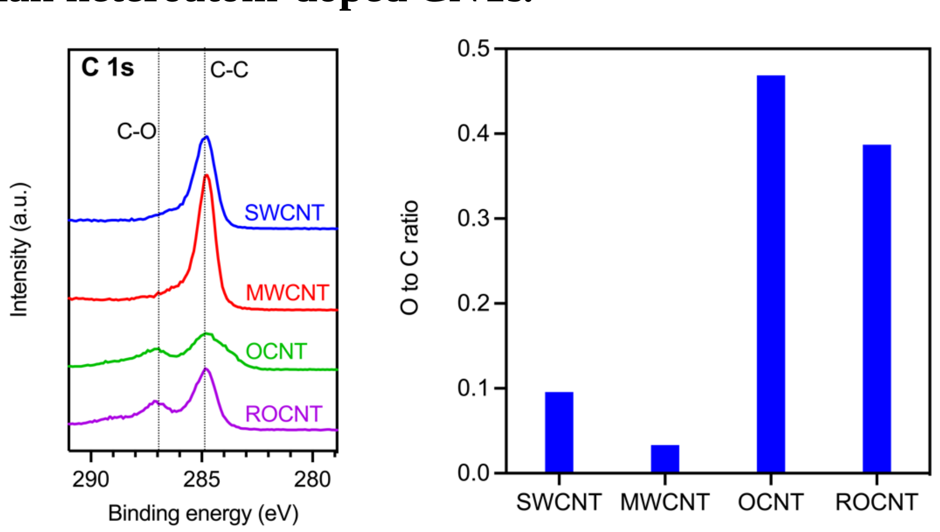
CNTs typically serve as a high surface area catalyst support for molecular catalysts and other nanomaterials.



Structural characterization

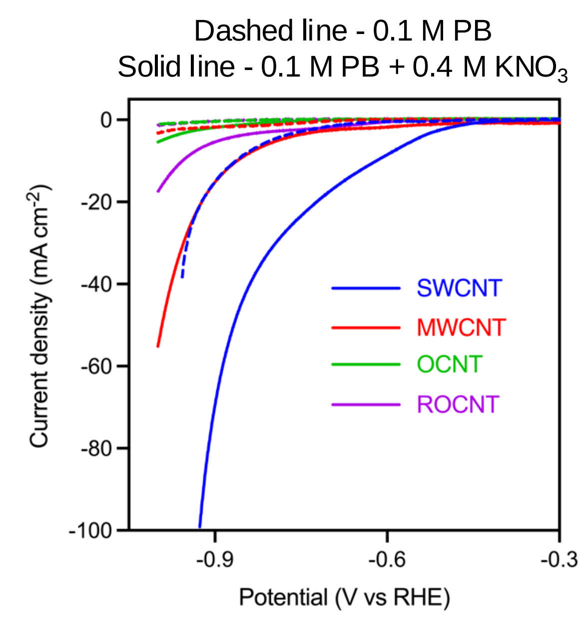


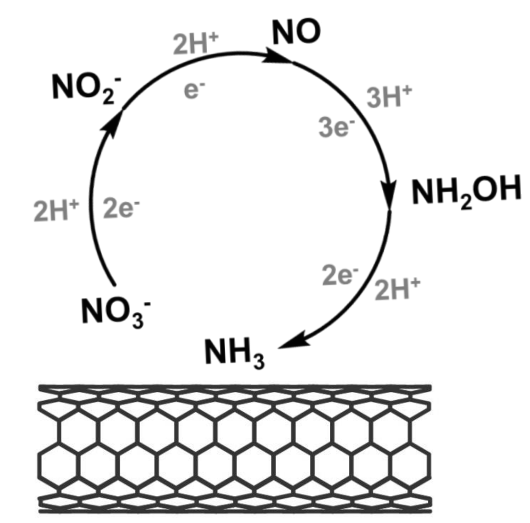
Pristine CNTs have less disorder and non-uniformity than heteroatom-doped CNTs.



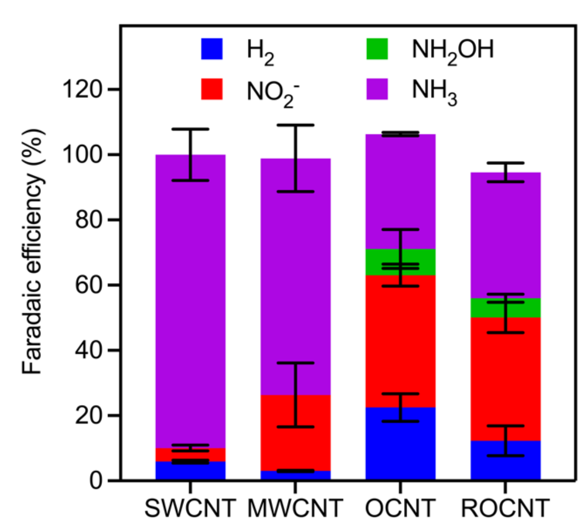
Partial restoration of the graphitic structure occurs when OCNTs are reduced to produce ROCNTs.

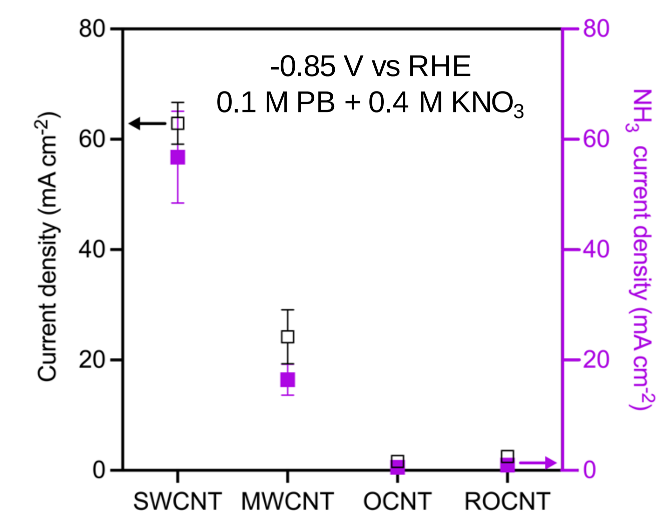
Catalytic performance of CNTs



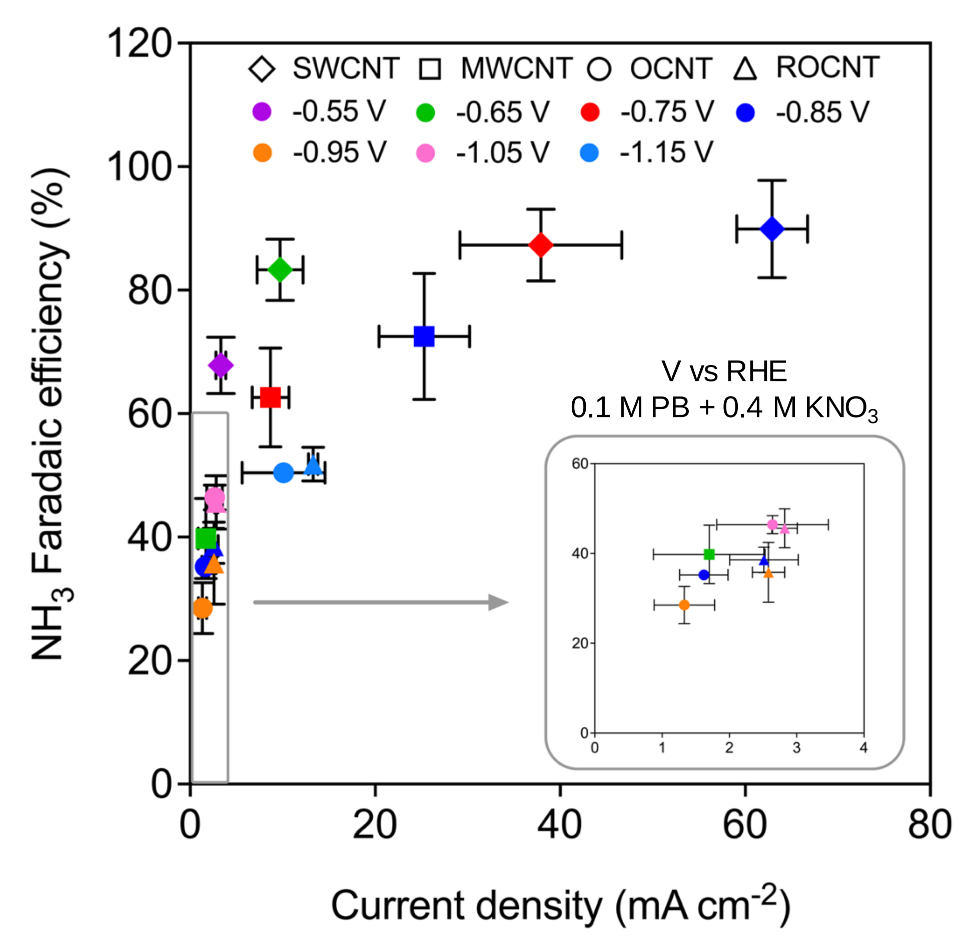


Both MWCNTs and SWCNTs are active and selective electrocatalysts for NH_3 production.



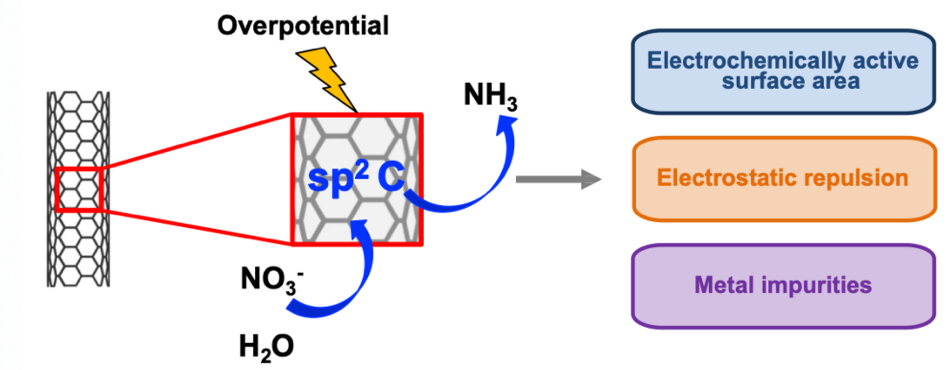


Heteroatom doping into CNTs lowers their catalytic activity.

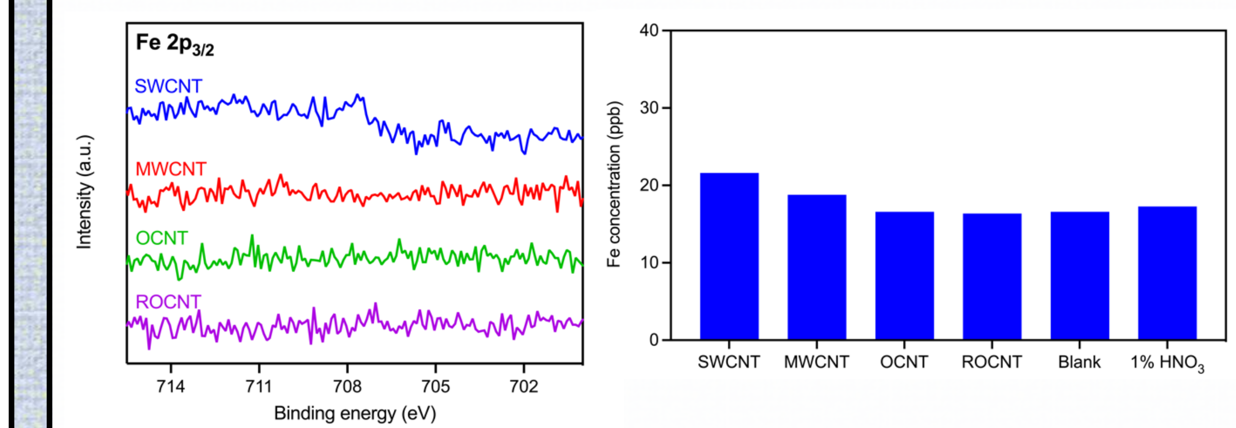


We find a positive correlation between NH_3 production and total current density, regardless of the type of CNT surface.

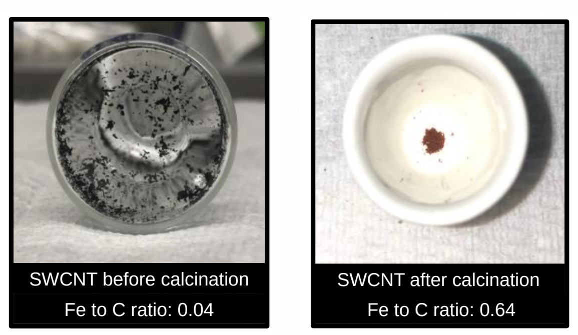
Origin of the catalytic performance of CNTs

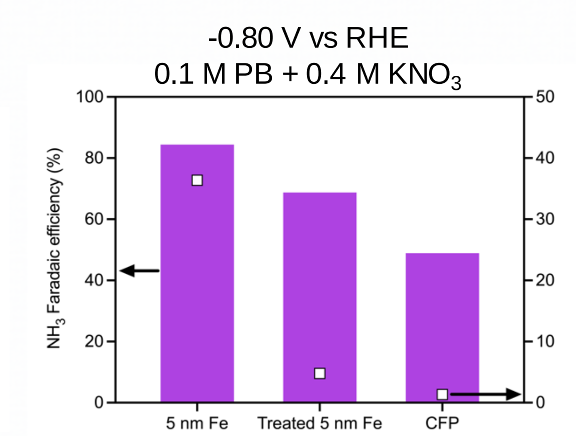


We confirm that doping MWCNTs with O and N functional groups does reduce the catalytic activity of the sp^2 C surface. At near-neutral pH, OCNTs have negative surface charges which are not present on MWCNTs.



SWCNTs still contain Fe residues from their synthesis even after purification.





Metallic Fe is considerably active for NO_3^- reduction.

Conclusions and Future work

- Pristine CNTs are active and selective electrocatalysts.
- Heteroatom doping decreases catalytic performance.
- Synthesize SWCNTs using a green synthesis procedure that utilizes biomass as a carbon source and catalyst to ensure that SWCNTs do not contain metal impurities.

Reference and acknowledgements

Harmon, N.J. *et al.* *ACS Catal.* **2022**, *12*, 9135– 9142.

Let's connect!
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